Atomic data of low-charged Sn ions for lithography applications

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Outline of talk

- Atomic Physics at Los Alamos National Laboratory (LANL)
  - Decades long research program at LANL including modern atomic structure & scattering codes
  - Opacity table production
    - Recent release of new LANL opacity tables for H through Zn
  - Comparisons of spectra with a variety of plasma physics experiments
- General considerations for Sn opacity & emissivity
  - Initial thought was to produce opacity tables for Sn for use by the (lithography) modeling community
  - Exploration started with atomic structure accuracy considerations: configuration-interaction vs intermediate-coupling
  - Completeness: how many configurations must we retain?
  - How does Sn opacity compare to previous work?
  - How do completeness/accuracy requirements vary with plasma conditions?
- Conclusions and what we have learned

Atomic Physics at Los Alamos National Laboratory

- LANL has maintained atomic structure and scattering computational tools for many decades
  - These have generally been motivated by 2 considerations:
  - Calculation of LTE opacity tables for wide ranges of materials & conditions
  - Calculation of spectra to compare with a variety of laser-produced and magnetically-confined plasmas; often these are in nonLTE
  - Often these considerations have focused on hot & dense plasmas
- LANL has developed (and continues to develop) a suite of atomic physics codes for this purpose
- We also develop the ATOMIC multi-purpose kinetics/opacity code to compare spectra and produce opacities over wide ranges of conditions

LANL structure codes: Fontes et al, JPB 48, 144014 (2015);
ATOMIC: Magee et al, 14th APS Topical Proceedings, 168 (2004);
Hakel et al, JQSRT 99, 265 (2006)

The LANL suite of atomic modeling codes

Atomic Physics Codes → Atomic Models → ATOMIC

CATS: Cowan Code

RATS: relativistic

ACE: e^- excitation

GIPPER: ionization

http://aphysics2.lanl.gov/tempweb

fine-structure
config-average
UTAs
MUTAs
energy levels
gf-values
e^- excitation
e^- ionization
photoionization
autoionization

LTE or NLTE
low or high-Z
populations

spectral modeling
emission
absorption
transmission
power loss

Recent LANL opacity tables

- Recent work by us has discussed the calculations of new LANL OPLIB opacity tables for H through Zn
- Data publicly available at:
  - ***http://aphysics2.lanl.gov/opacity/lanl***
- Calculations have been extensively compared with tables from other institutions and available experimental data
- Although new tables agree well with many measurements, the differences with the most recent Sandia Z-pinch measurements persist – an unresolved controversy in our field
- New opacities have recently been used in stellar modeling

Fe opacity

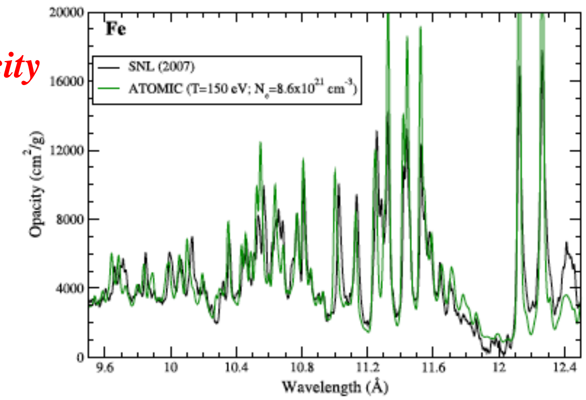


Figure 2. Comparison of Fe opacity measured in 2007 (Bailey et al. 2007) using the Sandia National Laboratory Z-pinch (black line) with an ATOMIC calculation at a temperature (T) of 150 eV and an electron density (N_e) of $8.6 \times 10^{21}\,\mathrm{cm}^{-3}$. The experimental conditions were inferred to be $T = 156 \pm 6$ eV and $N_e = 6.9 \times 10^{21}\,\mathrm{cm}^{-3}$ $\pm 25\%$ (Bailey et al. 2007).

Cr opacity

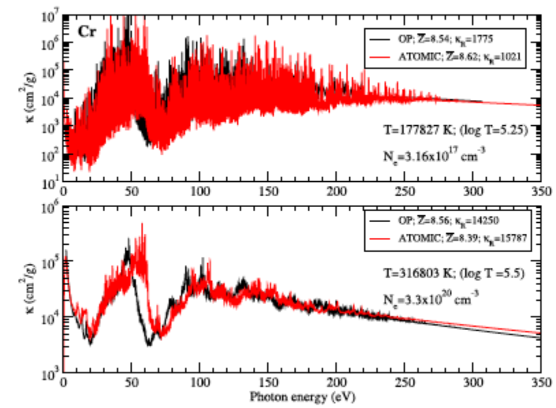
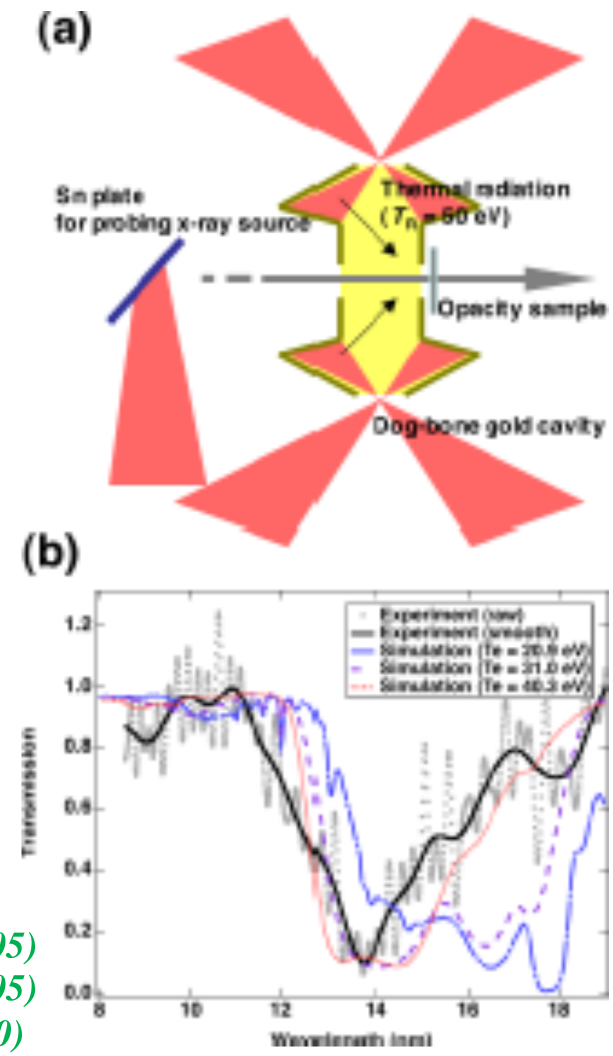


Figure 3. Comparison of the opacity of Cr from the OP database (Badnell et al. 2005; black lines) and the current ATOMIC calculations (red lines) at two different sets of temperatures (T) and electron densities (N_e) as indicated. The average ionization ($\bar{Z}$), and Rosseland mean opacity (κ_R), in cm^2/g, are indicated in the figure captions.

J. Colgan et al, Ap. J. 817, 116 (2016)

Why do we care about emission from tin?

- What has been done before on the experimental front?
- Measurements made using a ‘dog bone’ gold cavity and the Gekko laser facility (Japan) in 2005
- Note the strong dip in transmission (peak in opacity/emissivity) near 13.5 nm
- Measurements were ‘close’ to LTE and compared to calculations made using the HULLAC code
- Reasonable agreement was obtained but possible presence of temperature gradients was noted



Fujioka et al, PRL 95, 235004 (2005)
Lysaght et al, PRA 72, 014502 (2005)
Ohashi et al, JPB 43, 065204 (2010)

Why do we care about emission from tin?

- What has been done before on the theoretical front?
- Several groups have performed numerous studies of the emissivity of tin (eg UCD group/Chinese group/Japanese groups)
- The importance of accurate atomic structure is well known
- However, although a number of studies have been published, no comprehensive set of tables for opacity/emission seems to be available. It is not known how the radiative properties of tin change as plasma conditions vary
- We hope to address this by producing detailed opacity tables for tin over the temperature range of interest in lithography (0 to tens of eV)

Zeng et al, PRE 82, 026409 (2010)
Sasaki et al, JAP 107, 113303 (2010)
D'Arcy et al, JPB 42, 165207 (2009)
D'Arcy et al, PRA 79, 042509 (2009)
Churilov & Ryabtsev, Phys. Scr. 73, 614 (2006)

Sn opacity at low temperatures

- We construct a model with a number of Δn=0 permutations from the ground configuration
- We aim to account for sufficient **configuration-interaction (CI)** between the configurations that are retained
- We also run an **intermediate-coupling (IC)** calculation
- IC includes the spin-orbit coupling *between* the levels generated *within* one configuration
- **CI** includes the spin-orbit coupling and the 2-electron interactions *between* configurations
  - ***much*** more computationally expensive
- Both approaches predict the same number of fine-structure energy levels
- IC calculations generally used for opacity tables because one can easily include large numbers of configurations

Sn^{10+} as an example (40 electrons):

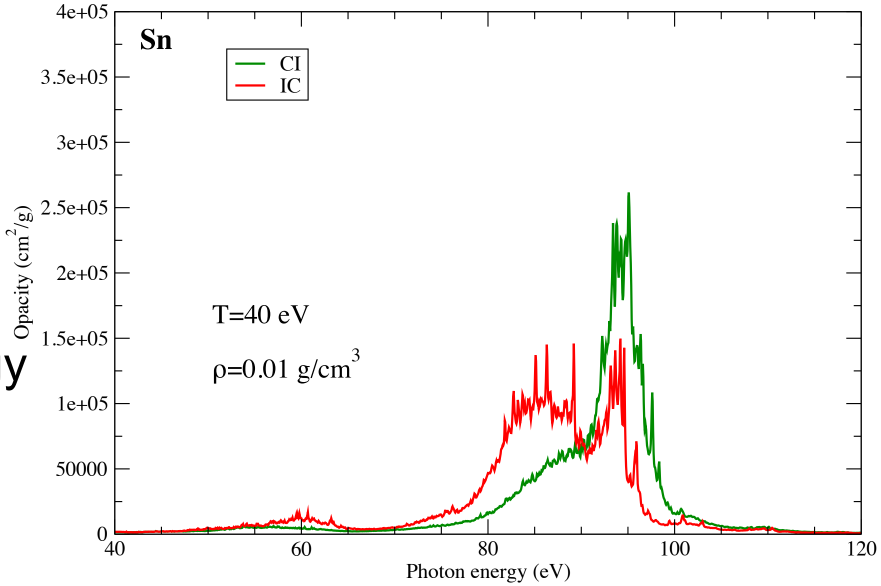
$4s^2 4p^6 4d^4$	$4s^2 4p^6 4d^3 (5\text{-}8)^1$
$4s^2 4p^6 4d^3 4f^1$	$4s^2 4p^6 4d^2 4f^1 (5\text{-}8)^1$
$4s^2 4p^6 4d^2 4f^2$	$4s^2 4p^5 4d^4 (5\text{-}8)^1$
$4s^2 4p^6 4d^1 4f^3$	$4s^2 4p^5 4d^3 4f^1 (5\text{-}8)^1$
$4s^2 4p^6 4f^4$	$4s^1 4p^6 4d^4 (5\text{-}8)^1$
$4s^2 4p^5 4d^5$	$4s^1 4p^6 4d^3 4f^1 (5\text{-}8)^1$
$4s^2 4p^5 4d^4 4f^1$	$4s^2 4p^4 4d^5 (5\text{-}8)^1$
$4s^2 4p^5 4d^3 4f^2$	$4s^2 4p^6 4d^2 5s^1 (6\text{-}8)^1$
$4s^1 4p^6 4d^5$	$4s^2 4p^6 4d^2 5p^1 (6\text{-}8)^1$
$4s^1 4p^6 4d^4 4f^1$	$4s^2 4p^6 4d^2 5d^1 (6\text{-}8)^1$
$4s^1 4p^6 4d^3 4f^2$	$4s^2 4p^5 4d^3 5s^1 (6\text{-}8)^1$
$4s^2 4p^4 4d^6$	
$4s^2 4p^4 4d^5 4f^1$	

182 configurations; > 330,000 levels

Sn opacity at 40 eV - importance of accuracy

- Sn opacity at 40 eV and density of 0.01 g/cc
- Substantial differences exist between IC and CI calculations
- Emissivity will closely resemble opacity over this limited energy range
- IC approximation produces opacity peaks over quite a wide photon energy range
- Inclusion of full CI produces a much narrower, more intense feature
  - This comes at a significant computational cost

Sn at 40 eV and 0.01 g/cc



Structure run-times:

IC=~ 3 days (serial code)

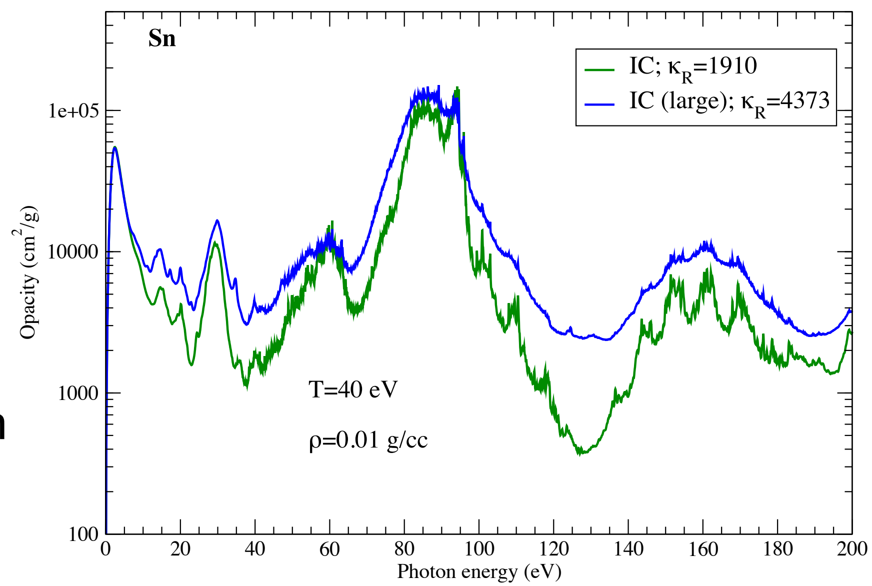
CI:~8 ***weeks*** *(parallel code)*

Ionicities 11-18 retained

Sn opacity at 40 eV - importance of completeness

- Sn opacity at 40 eV and density of 0.01 g/cc
- We consider only IC calculations
- Addition of many more configurations to the IC model can also change the opacity considerably
- The major peaks do not change much but the valleys between the peaks are filled in considerably by the absorption due to the extra configurations
- This has a significant effect on the Rosseland mean opacity
  - This quantity is a harmonic mean and so the absorption dips are relatively more important
- Emission may be less sensitive

Sn at 40 eV and 0.01 g/cc

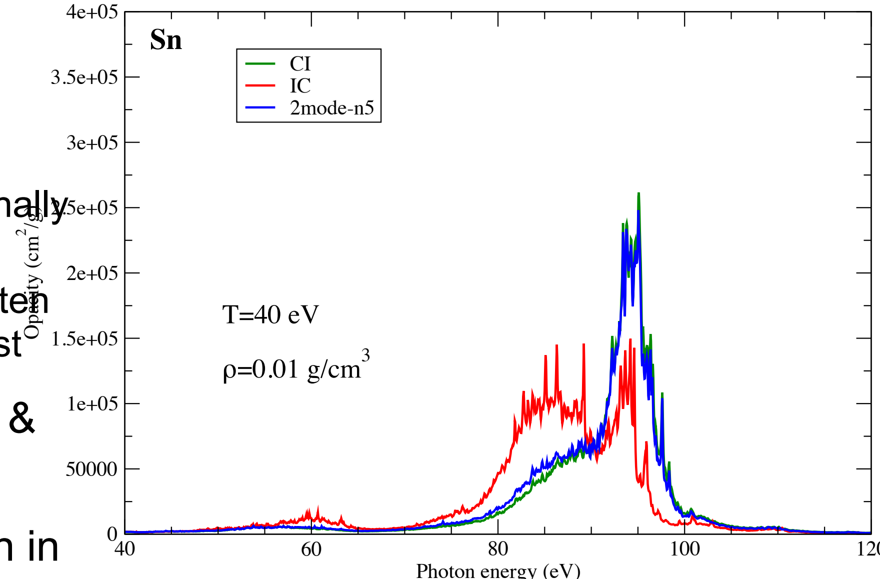


Ionicities 11-18 retained

Sn opacity at 40 eV – accuracy vs completeness

- In previous opacity tables a constant tension has been due to the competing requirements of accuracy & completeness
- Completeness was generally considered "more" important in terms of averaged quantities
  - Accuracy is always desirable but computationally very expensive
  - Improvements in accuracy also resulted in often marginal changes for most plasmas of interest
- However for Sn, it appears **BOTH** accuracy & completeness are required
- We developed a "**2mode**" type of calculation in which CI is only retained for $\Delta n=0$ and $\Delta n=1$ transitions; IC used for all other calculations
- The "**2mode**" calculation can almost perfectly reproduce the CI calculations with a fraction of the computational cost – and allows systematic inclusion of many more configurations

Sn at 40 eV and 0.01 g/cc

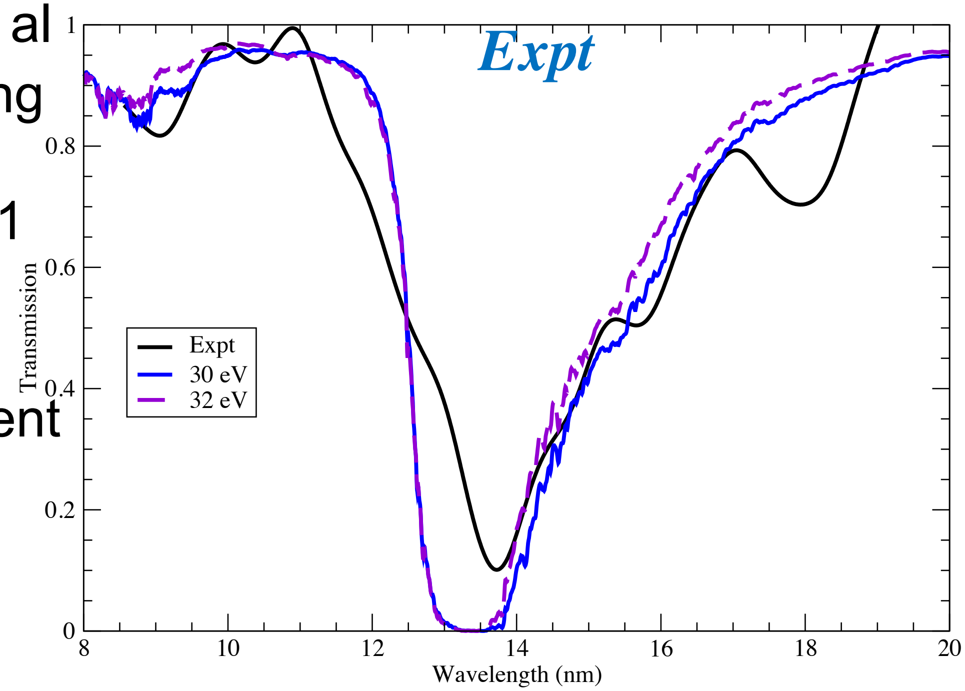


Structure run-times:

IC=~ 3 days (serial code)
*CI:~8 **weeks** (parallel code)*
2mode: 1 week (serial code)
2mode: 1 day (parallel code)

Sn opacity: comparison with experiment

- We are now in a position to compare with the measurements of Fujioka et al
  - Hydro simulations presented along with measurement indicated conditions of T=30 eV and ρ=0.01 g/cc
  - ATOMIC 2-mode calculation is in good agreement with measurement
    - Agreement worsens at lower wavelengths
  - Possible presence of inhomogeneities in plasma (temperature gradients) may complicate further analysis

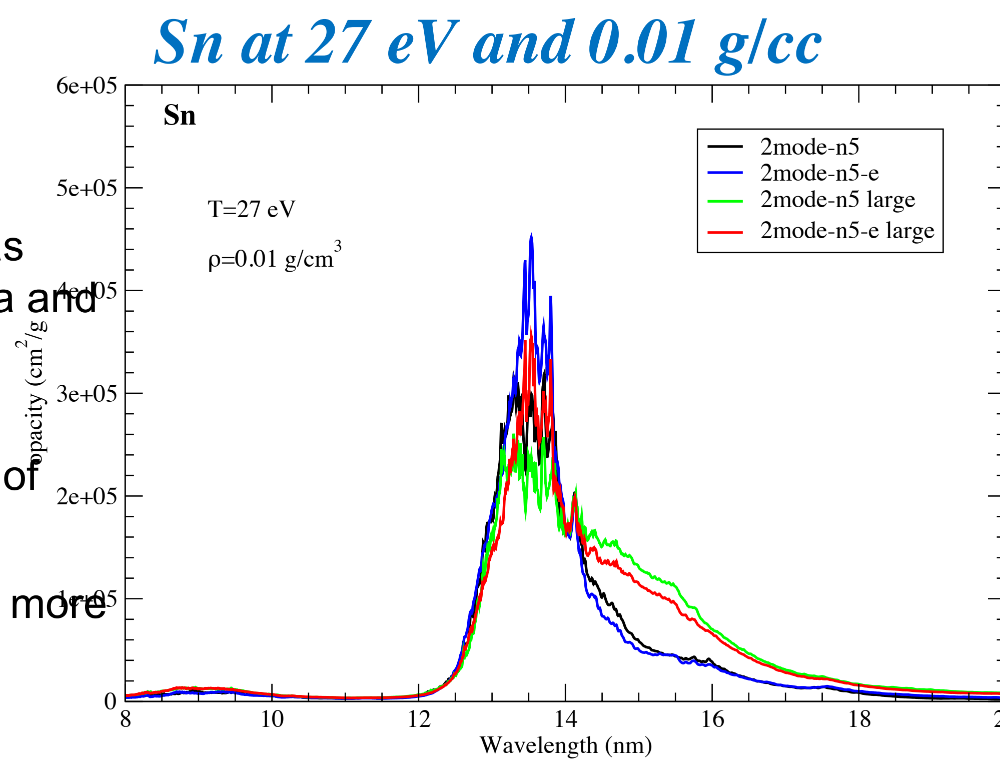


ATOMIC

Fujioka et al, PRL 95, 235004 (2005); Colgan et al, HEDP 23, 133 (2017)

Sn opacity at 27 eV and 0.01 g/cc

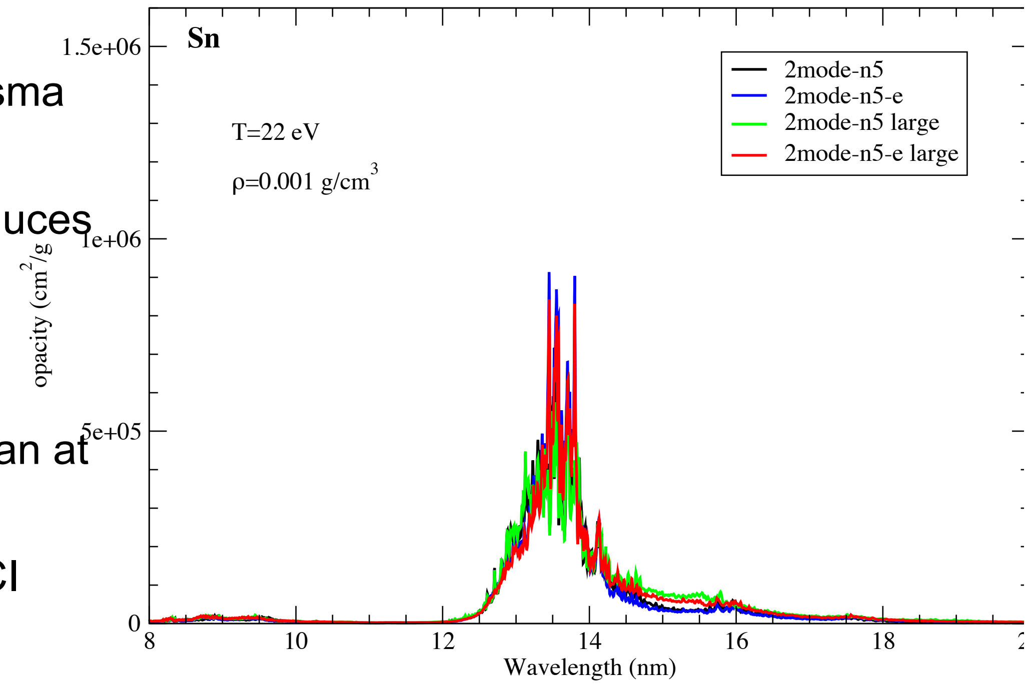
- *How can we assess convergence in our calculations?*
- *How does convergence depend on plasma conditions?*
- The "2mode-n5" model presented earlier was extended to include higher angular momenta and some more selected multiply excited configurations – "2mode-n5-e"
- Extended model **further** increases intensity of 13.5 nm intense feature
- Larger models (that incorporate significantly more configurations) also increase the opacity at wavelengths beyond 14 nm
- These extra configurations included using IC; however some may require inclusion in the CI portion of the calculation.
  - Such calculations are time-consuming but underway



Sn opacity at 22 eV and 0.001 g/cc

- *We consider lower densities, and also lower our temperature to keep roughly the same ion stage distribution*
- Does convergence change at different plasma conditions?
- CO_2 laser-produced plasma generally produces lower density plasma than (say) Japanese GEKKO laser experiment
- At lower densities we find that the extra configurations make less of a difference than at higher densities
- This is true for extra configurations in the CI calculation AND in the IC calculation
- Indicates convergence may be easier to reach at lower densities

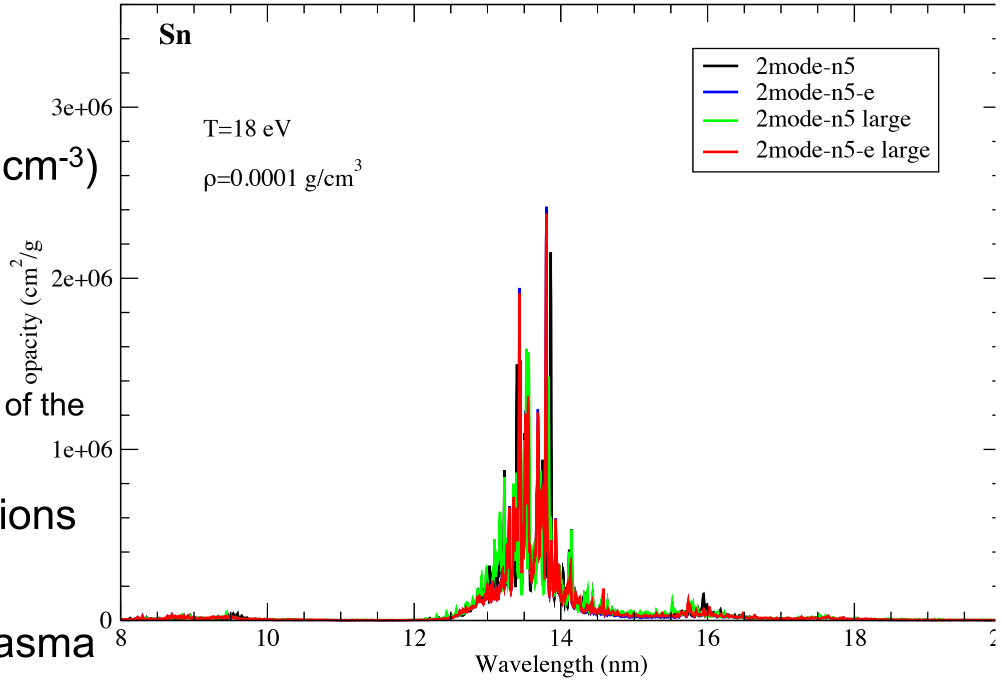
Sn at 22 eV and 0.001 g/cc



Sn opacity at 18 eV and 0.0001 g/cc

- *How can we assess convergence in our calculations?*
- *How does convergence depend on plasma conditions?*
- At lower densities of 0.1 mg/cc ($N_e \sim 5\text{x}10^{18}$ cm^{-3}) the lines become much narrower and the prominent features line up well from all calculations
  - These calculations were made with some artificial instrumental broadening added to reduce the density of the "forest" of line structure to make comparisons easier
- Indicates that our 2mode-n5-e large calculations may contain sufficient configurations
- On the other hand, at lower densities, the plasma is more likely to be in nonLTE, which introduces further complications
  - NonLTE calculations are significantly more time-consuming than the LTE calculations presented here

Sn at 18 eV and 0.0001 g/cc



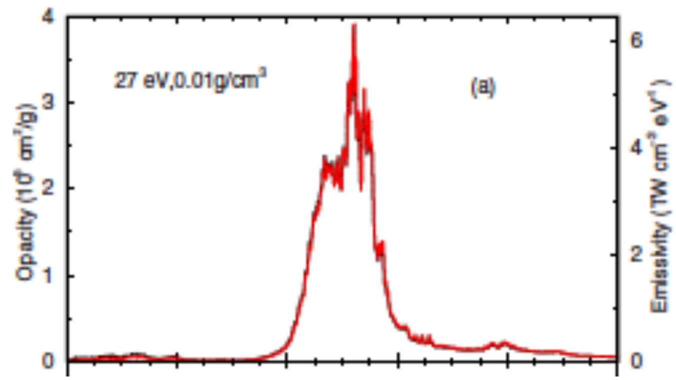
Conclusions & Future Work

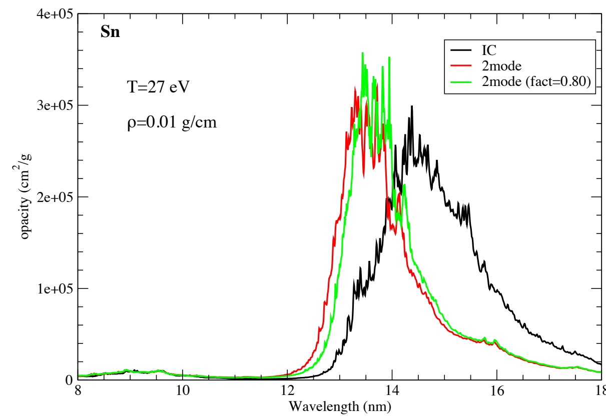
- Large-scale atomic data calculations have been performed for Sn ions of interest
- A "2-mode" approach that includes full configuration-interaction as well as contributions from other configurations via intermediate-coupling has been tested
- For the opacity of Sn, both **accuracy** AND **completeness** appear crucial in obtaining converged results at moderate densities
  - At lower densities, convergence with respect to number of configurations appears easier to obtain
- This underscores the difficulty in accurately modeling the spectra from such complex ions
- We hope our data can be useful to in-line nonLTE modeling efforts
- We continue with our efforts to produce tables of Sn opacities

Extra Slides

Sn opacity at 27 eV and 0.01 g/cc

- We can also examine the affect of modifying the (semi-empirical) scale factors in Cowan's code/ CATS
  - These factors scale the single-configuration Coulomb radial integrals within CATS
  - We reduce these from ~0.87 to ~0.80 to gauge the sensitivity of the resulting opacity
  - Modifying the scale factors produces a small but noticeable shift in the main absorption feature
  - It would be



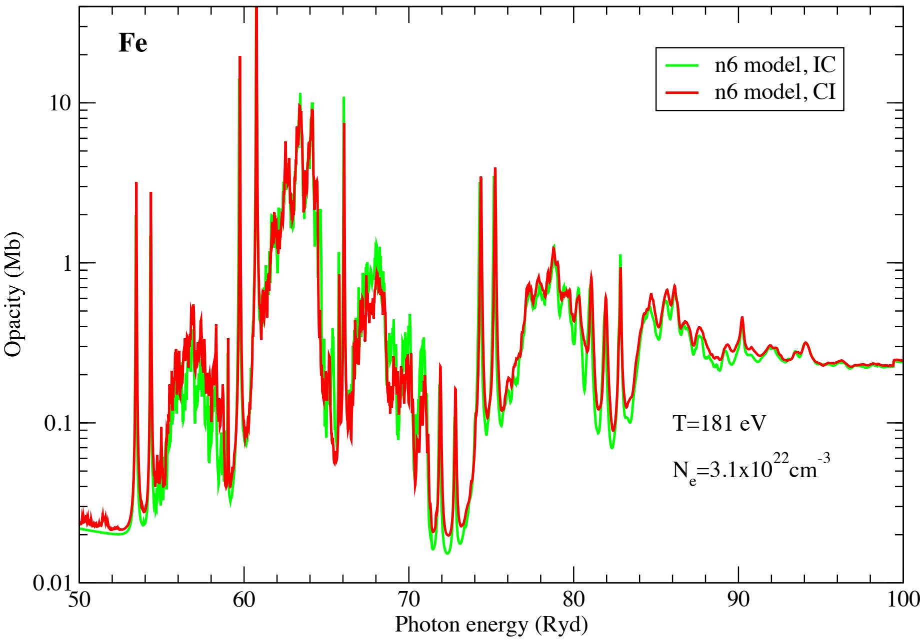


F ... ty for Sn p ... m^3 in a wa ... line) and (b) separa ... tion stages: The emissivity (in unit of 1 TW$=1\times10^{12}$ W) is given in a dotted red line.

Does this study have implications for our lower-Z opacity tables (eg Fe?)

- At higher temperatures (~200 eV) the inclusion of CI has an even smaller effect on the opacity
- No noticeable shift in line positions or even in strength of the major features
- Inclusion of CI not thought to be a factor in disagreement with the Fe SNL measurements

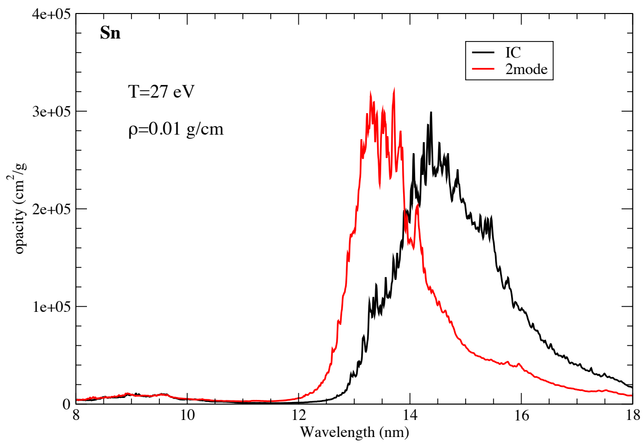
Fe opacity at 181 eV and N_e $3.1x10^{22}/cm^3$



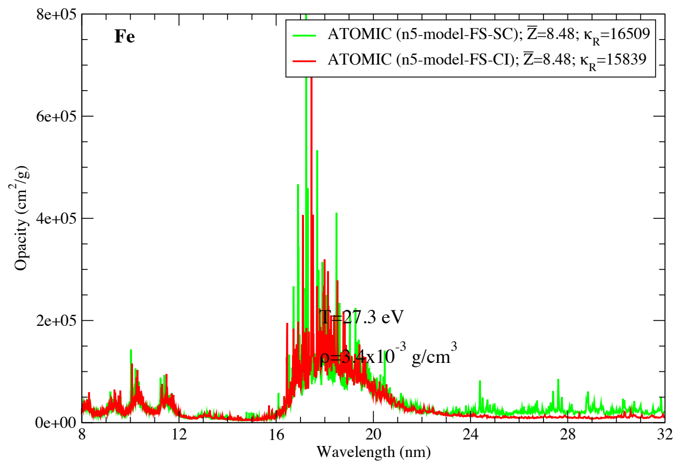
Inclusion of CI over IC has a relatively small effect

So why is CI relatively much more important in Sn compared to Fe?

Sn opacity at 27 eV and 0.01 g/cm³



Fe opacity at 27.3 eV and 3.4 mg/cm³



- Why does CI (compared to IC) seem to change the opacity of Sn so much more than the opacity of Fe?
- This may be because the transitions of importance in the Sn case are 4d-4f transitions; the effect of CI is very strong on these $\Delta n=0$ transitions involving larger units of angular momentum
- In the Fe case the transitions are 3p-3d; CI seems to have a smaller effect